

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Application of Hideaki Katsuta et al.

Serial No. 10/630,813

Filed: July 31, 2003

Art Unit: 1711

Examiner: Rachel F. Gorr

For: Light-Colored Water Based Intercoat Coating

Composition and Multi-Layer Coating Film Formed by  
Use of the Same

DECLARATION UNDER RULE 37 C.F.R. 1.132

HONORABLE COMMISSIONER OF PATENTS AND TRADEMARKS

WASHINGTON, D.C. 20231

Sir:

Now comes Hideaki Katsuta, who deposes and says that:

1. I am one of the inventors of the present  
invention of this patent application.

2. The following experiments shown by Experiments 1-  
8 were carried out by me or my direct supervision and  
control.

3. Experiments 1-8 duplicated Examples 1-8 of the  
present application respectively, except that in addition  
to anti-chipping properties test in the present application,  
i.e. anti-chipping properties test (A), a multi-layer  
coating film test panel was further subjected to anti-  
chipping properties test (B) as follows. Results are shown  
in Table I.

#### Preparation Example of Polyester Resin No. 1

A four-necked flask equipped with a heater, stirrer, thermometer, reflux condenser and a water separator was charged with 61.9 parts of 1,3-cyclohexane dicarboxylic acid, 70.1 parts of adipic acid, 62.8 parts of trimethylolpropane, 24.2 parts of neopentyl glycol and 44.6 parts of 1,4-cyclohexane dimethanol, followed by heating the content up from 160°C to 230°C over 3 hours, keeping at 230°C for one hour, distilling off a resulting condensation water by use of a fractionating column, adding 15.0 parts of anhydrous trimellitic acid to the resulting product, desolvating, neutralizing with dimethylethanolamine, and mixing with water to obtain polyester resin No. 1 having a solid content of 40%, hydroxyl value of 150 mgKOH/g, acid value of 35 mgKOH/g and a number average molecular weight of 2,000.

#### Preparation Example of Polyester Resin No. 2

A four-necked flask equipped with a heater, stirrer, thermometer, reflux condenser and a water separator was charged with 61.9 parts of 1,3-cyclohexane dicarboxylic acid, 70.1 parts of adipic acid, 62.8 parts of trimethylolpropane, 24.2 parts of neopentyl glycol and 44.6 parts of 1,4-cyclohexane dimethanol, followed by heating the content up from 160°C to 230°C over 3 hours, keeping at 230°C for one hour, distilling off a resulting condensation water by use of a fractionating column, adding 10.6 parts of anhydrous trimellitic acid to the resulting product,

desolvating, neutralizing with dimethylethanolamine, and mixing with water to obtain polyester resin No. 2 having a solid content of 40%, hydroxyl value of 168 mgKOH/g, acid value of 25 mgKOH/g and a number average molecular weight of 2,000.

#### Preparation Example of Polyester Resin No. 3

A four-necked flask equipped with a heater, stirrer, thermometer, reflux condenser and a water separator was charged with 43.8 parts of adipic acid, 77.0 parts of isophthalic acid, 54.6 parts of trimethylolpropane, 64.0 parts of butylethylpropanediol and 21.0 parts of neopentyl glycol, followed by reacting, adding 15.3 parts of anhydrous trimellitic acid to the reaction product, desolvating, neutralizing with dimethylethanolamine, and mixing with water to obtain polyester resin No. 3 having a solid content of 40%, hydroxyl value of 140 mgKOH/g, acid value of 35 mgKOH/g and a number average molecular weight of 2,000.

#### Preparation Example of Polyester Resin No. 4

A four-necked flask equipped with a heater, stirrer, thermometer, reflux condenser, nitrogen gas-introducing device and dropping device was charged with 182 parts of 1,6-hexanediol, 144 parts of trimethylol propane, 104 parts of Cardura E10 (trade name, marketed by Japan Epoxy Resins Co., Ltd., versatic acid glycidyl ester), 132 parts of adipic acid, and 246 parts of phthalic acid, followed by condensating at 230°C, cooling at the time when an acid

value became 2 or less, adding 46 parts of anhydrous trimellitic acid, reacting at 170°C for 30 minutes, adding dipropylene glycol monomethyl ether, cooling down to 80°C, neutralizing with dimethylethanolamine while stirring, and adding deionized water and dispersing to obtain an aqueous polyester resin solution having a solid content of 45%, an acid value of 35 mgKOH/g, a hydroxy value of 120 mgKOH/g and pH of 7.2.

Preparation Example of Curing Agent No. 1

The following composition containing components (1) to (6) was used to obtain the curing agent No. 1 as follows.  
Composition:

	<u>% by weight</u>
(1) 1,6-hexamethylene diisocyanate trimer	30
(2) polyoxyalkyleneamine (Jeffermine M-1000, trade name, marketed by Huntsman Chemical Co., Ltd.)	3
(3) 3,5-dimethylpyrazole (blocking agent)	16
(4) butyl glycol ether (solvent)	7
(5) sodium sulfosuccinate	4
(6) water	40

A reactor was charged with component (1), followed by slowly adding component (2), heating up to 60 to 70°C so that an isocyanate content may reach 20 to 21 mol%, keeping at 60 to 70°C until the isocyanate content reaches zero, while slowly adding component (3), adding components (4) and (5), mixing for 5 minutes, adding component (6) water, and forming a dispersion by use of a high speed mixer.

Jeffermine M-1000 is an active hydrogen-containing compound which reacts with HMDI (1,6-hexamethylene diisocyanate) trimer to form a polyisocyanate prepolymer. The sodium sulfosuccinate is an ionic surface active agent which assists controlling a particle size of the dispersion.

Experiment 1

A mixture of 112.5 parts (solid content 45 parts) of polyester resin No. 1, 110 parts (solid content 55 parts) of curing agent No. 1 and 80 parts (solid content 80 parts) of JR-806 (Note 2) was dispersed, followed by adding deionized water and controlling a viscosity at 50 seconds, 20° C, Ford-Cup No. 4 to obtain a water based intercoat coating composition No. 1. Results are shown in Table I.

#### Experiments 2-8

Experiment 1 was duplicated according to the formulations in Table I to obtain water based intercoat coating compositions No. 2 to No. 8 of Experiments 2-8 respectively. In Table I, a mixing amount is represented by solid content part by weight.

(Note 1) Urethane Emulsion (trade name, marketed by Sanyo Chemical Industries Ltd.).

(Note 2) JR 806: trade name, marketed by Tayka Corporation, titanium white.

(Note A) Byhydrol PT241, trade name, marketed by Sumitomo Bayel Urethane Co., Ltd., urethane-modified polyester resin.

Table I

		Experiments							
		1	2	3	4	5	6	7	8
Water based intercoat coating composition		1	2	3	4	5	6	7	8
Polyester resin No. 1		45		27	20				
Polyester resin No. 2			45						
Polyester resin No. 3						45			
Polyester resin No. 4							45		
Byhydrol PT241 (Note A)								30	
Acrylic resin					25				45
Curing agent No. 1		55	55	33	55	55	55	70	55
Curing agent No. 2									
Curing agent No. 3									
Curing agent No. 4									
UX 8100 (Note 1)				40					
JR 806 (Note 2)		80	80	80	80	80	80	80	80
Multi-layer coating film	anti-chipping properties test (A)	○	○	◎	○	○	○	○	○
	anti-chipping properties test (B)	◎	◎	◎	◎	○	○	○	○

Anti-chipping properties test (A): A test panel was mounted on a test panel holder of a JA-400 type rebounding gravels test instrument marketed by Saga test instruments Co., Ltd. (chipping test apparatus), followed by spraying 50 g of granite gravels having a particle size of No. 7 onto the surface of a coating film under an air pressure of 0.392 MPa (4 kgf/cm<sup>2</sup>) at -20°C, and visually evaluating degree of development of mars on the coating film as follows.

◎: Size of the mar is considerably small and the

intercoat coating film is slightly exposed,

○: Size of the mar is small and the intercoat coating film is exposed,

△: Size of the mar is small, but steel sheet substrate is exposed,

×: Size of the mar is considerably large, and the steel sheet substrate is largely exposed.

Anti-chipping properties test (B): The anti-chipping properties test (A) was duplicated except that an air pressure of 0.294 MPa (3 kgf/cm<sup>2</sup>) was used in place of the air pressure of 0.392 MPa (4 kgf/cm<sup>2</sup>), followed by visually evaluating degree of development of mars on the coating film as follows.

◎: Size of the mar is considerably small and the intercoat coating film is slightly exposed,

○: Size of the mar is small and the intercoat coating film is exposed,

△: Size of the mar is small, but steel sheet substrate is exposed,

×: Size of the mar is considerably large, and the steel sheet substrate is largely exposed.

#### Conclusion:

According to the presently claimed invention, the use of the specified hydroxyl group-containing polyester resin (A<sub>2</sub>) containing the alicyclic polybasic acid (a<sub>1</sub>) and/or the alicyclic polyhydric alcohol (a<sub>2</sub>) makes it possible to

particularly improve anti-chipping properties compared with hydroxyl group-containing polyester resins No. 3 and No. 4, and with urethane-modified hydroxyl group-containing polyester resin (Byhydrol PT241), which do not contain the alicyclic polybasic acid ( $a_1$ ) and/or the alicyclic polyhydric alcohol ( $a_2$ ) as evident from comparison between the results of Experiment Nos. 1-4 and those of Experiment Nos. 5-7.

4. The undersigned Petitioner declares further that all statements made herein of his own knowledge are true and that all statements made in information and belief are believed to be true, and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

5. Further, Deponent saith not.

Date:

12/24 /2004

Hideaki Katsuta

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